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Polyester Resin Composition and Sheet Production Method

Your Ref: 103003 - 05

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(54) [Title of the Invention] Polyester Resin Composition and Sheet Production Method

(57) [Summary]

[Object] To provide a polyester resin composition primarily consisting of a specific noncrystalline or low-crystalline polyester resin, which is formulated with specific resins and compounds to achieve excellent release properties from calender rollers, as well as solvent resistance and resistance to the heat of embossing.

[Means of Achievement] A polyester resin composition characterized in that 0.1-3 parts by weight of an organophosphoric acid triester compound as a lubricant are added per 100 parts by weight of a polyester resin component comprising 60-95% by weight of a noncrystalline or low-crystalline polyester resin composed of a dicarboxylic acid component, principally consisting of terephthalic acid, and a diol component composed of 15-80 mol% of 1,4-cyclohexane

dimethanol and 20-85 mol% of ethylene glycol; and 5-40% by weight of at least one type of highly crystalline polyester selected from polybutylene terephthalate resin and polyethylene terephthalate resin.

[Claims]

[Claim 1] A polyester resin composition, characterized in that 0.1-3 parts by weight of an organophosphoric acid triester compound as a lubricant are added per 100 parts by weight of a polyester resin component comprising 60-95% by weight of a noncrystalline or low-crystalline polyester resin composed of a dicarboxylic acid component, principally consisting of terephthalic acid, and a diol component composed of 15-80 mol% of 1,4-cyclohexane dimethanol and 20-85 mol% of ethylene glycol; and 5-40% by weight of at least one type of highly crystalline polyester selected from polybutylene terephthalate resin and polyethylene terephthalate resin.

[Claim 2] The polyester resin composition according to Claim 1, characterized in that a core-shell modifier prepared by means of the graft polymerization of a vinyl monomer shell onto a core consisting of a rubber component is further compounded in an amount of 1-20 parts by weight per 100 parts by weight of the polyester resin component.

[Claim 3] The polyester resin composition according to Claim 2, characterized in that the core component of the core-shell modifier is acrylic rubber, olefin rubber, diene rubber, or acrylic-siloxane compound rubber.

[Claim 4] The polyester resin composition according to Claims 1 through 3, characterized by being used for calendering applications.

[Detailed Description of the Invention]

[0001]

[Technological Field of the Invention] The present invention relates to a polyester composition having adequate molten processing capabilities when used in calendering and extrusion molding processes; more specifically, it relates to a polyester resin composition having excellent properties such as release from calender rollers, transparency, resistance to heat from embossing, and solvent resistance.

[0002]

[Prior Art] Polyester resin sheets enjoy widespread usage as surface coverings or wood finishes in furniture, utensils, doors, shelf boards, and the like; decorative sheets for cosmetic surface treatments in light electrical appliance products such as refrigerators and television cabinets; cabinets for use in the office; and packaging material for foodstuffs, medical equipment, etc. It was generally the case in the prior art that such polyester resin sheets were produced by means of melting resin pellets and molding them into sheets by means of extrusion. However, such extrusion molding processes involved the use of a T-die to extrude the sheets to a specific thickness, and the sheets were then processed with a casting roller whose temperature was kept at or below the glass transition temperature. The speed with which the sheets were molded was low, and the productivity could not be regarded as satisfactory.

[0003] The use of calender processing on such polyester resins to produce sheets, or the use of secondary processing such as heat lamination or heat embossing of the sheets was plagued by the strong adhesion of the sheets to the hot rollers. Japanese Unexamined Patent Publication (Kokai) 11-343353 disclosed the use of a fatty acid ester type lubricant to inhibit adhesion and to make calendering possible during the molding of sheets by means of calendering. However, even with the use of a fatty acid ester lubricant, the melt viscosity remained so high as to make it difficult produce comparatively thin sheets of 0.05 - 0.2 mm or to perform secondary processing such as high-temperature embossing.

[0004]

[Problems to Be Solved by the Invention] The objective of the present invention is to provide a polyester resin composition that provides easy release from calender rollers, resistance to the heat of embossing, and solvent resistance by means of formulating specific resins and compounds in a polyester resin composition primarily consisting of a specific noncrystalline or low-crystalline polyester resin.

[0005]

[Means Used to Solve the Above-Mentioned Problems] The present invention provides a polyester resin composition characterized in that 0.1-3 parts by weight of an organophosphoric acid triester compound as a lubricant are added per 100 parts by weight of a polyester resin

component comprising 60-95% by weight of a noncrystalline or low-crystalline polyester resin composed of a dicarboxylic acid component, principally consisting of terephthalic acid, and a diol component composed of 15-80 mol% of 1,4-cyclohexane dimethanol and 20-85 mol% of ethylene glycol; and 5-40% by weight of at least one type of highly crystalline polyester selected from polybutylene terephthalate resin and polyethylene terephthalate resin.

[0006] In cases where transparency is not required, the polyester resin composition according to Claim 1 may be characterized in that a core-shell modifier prepared by means of the graft polymerization of a vinyl monomer shell onto a core consisting of a rubber component is further compounded in an amount of 1-20 parts by weight per 100 parts by weight of the polyester resin component.

[0007]

[Embodiments of the Invention] The present invention shall be explained in detail below. The noncrystalline or low-crystalline polyester resin of this invention is composed of a dicarboxylic acid component principally consisting of terephthalic acid, and a diol component consisting of 15-80 mol% of 1,4-cyclohexane dimethanol and 20-85 mol% of ethylene glycol. Specific examples include noncrystalline polyester resin with approximately 30 mol% of 1,4-cyclohexane dimethanol such as "PETG 6763" and "Provista" made by Eastman Chemical Company, or low-crystalline polyester with approximately 70 mol% of 1,4-cyclohexane dimethanol such as "PCTG" and "PCTA" made by Eastman Chemical Company.

[0008] In the present invention, a highly crystalline polyester resin is compounded in addition to the foregoing noncrystalline/low-crystalline polyester resin. Specific examples include polybutylene terephthalate resin and polyethylene terephthalate resin. The highly crystalline polyester resin can be compounded in a ratio of 5-40% by weight in relation to the polyester resin component comprising the noncrystalline or low-crystalline polyester resin and the highly crystalline polyester resin. Should less than 5% by weight of the highly crystalline polyester resin be compounded, solvent resistance will be inadequate, whereas should 40% by weight be exceeded, reheating of the sheet will cause crystallization and the loss of transparency, an undesirable decline in the melt viscosity, and adverse effects on the high temperature embossing characteristics.

[0009] In the present invention, 0.1-3 parts by weight, and preferably 0.3-2.5 parts by weight, of an organophosphoric acid triester compound as a lubricant are compounded per 100 parts of the foregoing polyester resin component. This organic triester compound may be expressed by the general formula

$$[RO\ (CH_2\ CH_2\ O)_m]_3\ PO$$

(where R represents the same or different alkyl group or alkylaryl group with 10-22 carbon atoms, and m is an integer of 1-10). Should less than 0.1 part by weight be compounded, calendering properties and working properties during high-temperature embossing will decline, whereas if 3 parts by weight is exceeded, transparency will worsen, and printing properties will be adversely affected by large numbers of eruptions on the sheet surface.

[0010] A core-shell modifier prepared by means of the graft polymerization of a vinyl monomer shell onto a core consisting of a rubber component may be compounded in an amount of 1-20 parts by weight per 100 parts by weight of the foregoing polyester resin component as an optionally compounded core-shell modifying agent. The rubber component of the core-shell modifier may consist of acrylic rubber, olefin rubber, diene rubber, acryl-siloxane compound rubber, etc. A typical example of an acrylic rubber core-shell modifier is a modifier in which a vinyl monomer such as methyl methacrylate, acrylonitrile, or styrene is graft polymerized to form a shell on a core composed of acrylic rubber prepared by means of polymerizing a small amount of a crosslinking monomer with an acrylic acid ester such as butyl acrylate. Commercially available resins suited to this application include "HIA-80" made by Kureha Chemical Industry Co., Ltd., "Acryloid KM-330" made by Rohm and Haas, and "Metablen W-300" and Metablen W-530 made by Mitsubishi Rayon Co., Ltd. Also, acryl-siloxane compound rubber core-shell modifiers may be prepared by means of a process in which the foregoing vinyl monomer is graft polymerized upon a core of a compound rubber consisting of an acrylic polymer and a polyorganosiloxane. These modifiers are commercially available as "Metablen S200" and "Metablen SX006" made by Mitsubishi Rayon Co., Ltd. Such core-shell modifiers can contribute to lubrication of the resin composition and can raise its melt viscosity, and are effective for making it easier to transport the composition in a heated and gelled state, increasing the precision with which the materials can be fed to the calender rollers in predetermined

amounts, improving the precision in sheet thickness, and allowing sheets having excellent smoothness to be produced.

[0011] Acrylic processing aids or lubricants other than the foregoing organophosphoric acid triester compounds may be compounded as needed so long as they do not detract from the properties delivered by means of the present invention. Preferred examples of such acrylic processing aids include copolymers with a weight-average molecular weight ranging from 1 million to 5 million, which are primarily composed of acrylic acid esters, including acrylic acid, methyl acrylate, ethyl acrylate, n-butyl acrylate, isobutyl acrylate, and 2-ethylhexyl acrylate; or methacrylic acid esters, including methyl methacrylate, ethyl methacrylate, n-butyl methacrylate, isobutyl methacrylate, 2-ethylhexyl methacrylate, etc. Examples include "Metablen P-551" and "Metablen P-530A" made by Mitsubishi Rayon Co., Ltd., and "Kane Ace PA30" and "Kane Ace PA-100" by Kanegafuchi Chemical Industries Co., Ltd.

[0012] Such acrylic processing aids can be compounded in an amount of 10 parts by weight or less, preferably 0.5-8 parts by weight, and even more preferably 1 to 6 parts by weight, per 100 parts by weight of the polyester resin composition. The compounding has the effect of increasing the lubricity and melt viscosity and producing thin uniform surfaces.

[0013] In the present invention, various conventional additives may also be compounded, including pigments, hindered amine light stabilizers, ultraviolet absorbers, antioxidants, antistatic agents, inorganic and organic fillers, and compatible resins.

[0014] The polyester resin composition of this invention is prepared by means of a process in which the resin components, organophosphoric acid triester compounds, and optional additives are compounded in the desired amounts, the compound is melted and kneaded to produce a starting material, and sheets are formed by means of the usual calendering. The calender roll temperature is usually set to 165-190°C, and is preferably 175-185°C.

[0015]

[Working Examples] The invention will be more specifically described below through the use of working examples and comparative examples, but the invention is not confined to these examples. The items evaluated and the evaluation methods were as follows.

[0016] (1) Solvent Resistance

Calendered sheets 0.3 mm thick were cut into 50-mm-wide and 150-mm-long pieces, and then the sheet surfaces were wiped in twenty back and forth passes with 1 cc of the solvents shown in Table 1 absorbed into a piece of defatted cotton (approximately 30 mm square, 170 mg). After being allowed to stand for 20 minutes, the sheets were visually inspected and evaluated as follows.

o: Sheet exhibited no external deformation

Δ: Sheet exhibited slight external deformation

X: Sheet was solvent saturated and exhibited significant external deformation

[0017] (2) High-Temperature Embossing Properties

The highest temperature that was maintained on the surface of an embossing roller and that allowed at least 80% of the embossing depth to be maintained was measured when a relief was transfer-molded with the aid of the embossing roller. The roller had projections and depressions with a depth of about 20 μm and had a surface temperature of 110 to 150°C.

*: 150°C or higher

o: 130°C or higher, less than 150°C

Δ: 110°C or higher, less than 130°C

X: less than 110°C

[0018] (3) Transparency (haze value)

The haze value was measured in accordance with JIS-K7105 for sheets that were 0.3 mm thick and were manufactured by means of calendering.

o: Haze value less than 5.0%

Δ: Haze value 5.0% or greater, less than 10.0%

X: Haze value 10.0% or greater

[0019] Working Example 1

One part by weight of an organophosphoric acid triester compound ("LTP-2" made by Kawaken Fine Chemicals Co., Ltd., (polyoxyethylene lauryl ether) phosphoric acid triester) as a lubricant and 1 part by weight of a phenolic antioxidant were compounded per 100 parts by weight of a polyester resin component comprising 90% by weight of a noncrystalline polyester resin ("PETG6763" made by Eastman Chemical Company) and 10% by weight of polybutylene terephthalate resin ("600FP" made by Polyplastics Co., Ltd.) as a highly crystalline polyester

resin. Conventional methods were used for calender processing to prepare a transparent polyester resin sheet that was 0.3 mm thick. Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties, and transparency appear in Table 1.

[0020] Working Example 2

One part by weight of an organophosphoric acid triester compound ("LTP-2" made by Kawaken Fine Chemicals Co., Ltd.) as a lubricant and 1 part by weight of a phenolic antioxidant were compounded per 100 parts by weight of a polyester resin component comprising 40% by weight of a noncrystalline polyester resin ("PETG6763" made by Eastman Chemical Company), 40% by weight of a low-crystalline polyester resin ("PCTG" made by Eastman Chemical Company), and 20% by weight of polybutylene terephthalate resin ("600FP" made by Polyplastics Co., Ltd.) as a highly crystalline polyester resin. Conventional calender processing was used to prepare a 0.3-mm-thick transparent polyester resin sheet. Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0021] Working Example 3

One part by weight of an organophosphoric acid triester compound ("LTP-2" made by Kawaken Fine Chemicals Co., Ltd.) as a lubricant, and 1 part by weight of a phenolic antioxidant were compounded per 100 parts by weight of a polyester resin component comprising 40% by weight of a noncrystalline polyester resin ("PETG6763" made by Eastman Chemical Company), 30% by weight of a low-crystalline polyester resin ("PCTG" made by Eastman Chemical Company), and 30% by weight of polybutylene terephthalate resin ("600FP" made by Kawaken Fine Chemicals Co., Ltd.) as a highly crystalline polyester resin. Conventional calender processing was used to prepare a 0.3-mm-thick transparent polyester resin sheet. Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0022] Working Example 4

One part by weight of an organophosphoric acid triester compound ("LTP-2" made by Kawaken Fine Chemicals Co., Ltd.) as a lubricant and 1 part by weight of a phenolic antioxidant were compounded per 100 parts by weight of a polyester resin component comprising 50% by weight of a noncrystalline polyester resin ("PETG6763" made by Eastman Chemical Company),

20% by weight of a low-crystalline polyester resin ("PCTA" made by Eastman Chemical Company), and 30% by weight of polyethylene terephthalate resin ("MA521H" made by Mitsubishi Rayon Co., Ltd.) as a highly crystalline polyester resin. Conventional calender processing was used to prepare a 0.3-mm-thick transparent polyester resin sheet. Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0023] Comparative Example 1

One part by weight of an organophosphoric acid triester compound ("LTP-2" made by Eastman Chemical Company) as a lubricant and 1 part by weight of a phenolic antioxidant were compounded per 100 parts by weight of a noncrystalline polyester resin ("PETG6763" made by Eastman Chemical Company). Conventional calender processing was used to prepare a 0.3-mm-thick transparent polyester resin sheet. Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0024] Comparative Example 2

A transparent polyester resin sheet was manufactured by means of the usual calendering process in the same manner as described in Comparative Example 1, except that the lubricant in Comparative Example 1 was substituted with 1 part of an organophosphoric acid diester lubricant compound ((polyoxyethylene tridecyl ether) phosphoric acid diester) made by Akishima Chemical Industries Co., Ltd.). Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0025] Comparative Example 3

A polyester resin sheet was manufactured by means of the usual calendering process in the same manner as described in Working Example 1, except that the lubricant in Working Example 1 was substituted with 1 part of montan wax (made by Clariant Japan). Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0026] Comparative Example 4

One part by weight of an organophosphoric acid triester compound ("LTP-2" made by Kawaken Fine Chemicals Co., Ltd.) and 1 part by weight of a phenolic antioxidant were

compounded per 100 parts by weight of a polyester resin component comprising 50% by weight of a noncrystalline polyester resin ("PETG6763" made be Eastman Chemical Company) and 50% by weight of polybutylene terephthalate resin ("600FP" made by Polyplastics Co., Ltd.) as a highly crystalline polyester resin. Conventional calender processing was used to prepare a 0.3-mm-thick transparent polyester resin sheet. Both thickness precision and calendering properties were good. The evaluations for solvent resistance, high temperature embossing properties and transparency appear in Table 1.

[0027]

Table 1

		Examples				Comparative Examples			
		1	2	3	4	1	2	3	4
Noncrystalline polyester resin •PET G6763		90	40	40	50	100	100	90	50
Low-crystalline polyester resin •PCTG		40	40	30					
•PCTA					30				
Highly crystalline polyester resin •600 FP		10	20	30				10	50
•MA521H					20				
Lubricant •Phosphoric acid triester		1	1	1	1	1			1
•Phosphoric acid diester							1		
•Montan wax								1	
Evaluation	Solvent Resistance •Ethyl acetate	o	o	o	o	X	X	o	o
	•Toluene	o	o	o	o	X	X	o	o
	•Methylethyl ketone	o	o	o	o	X	X	o	o
	High-temperature embossing properties	*	*	*	*	*	o	o	Δ
	Transparency	o	o	o	o	o	o	X	o

[0028]

[Effect of the Invention] Since the polyester resin composition of this invention provides good release from calender rollers, high temperature embossing properties, and resistance to solvents, its calendering properties are excellent and it may be used to manufacture decorative sheets, wrapping materials, and the like.